

AMENDMENTS TO THE CLAIMS

This listing of claims will replace all prior versions and listings of claims in the application:

LISTING OF CLAIMS:

1. (currently amended): An enzyme electrode comprising:
a ~~portion of an~~ electrode formed on an insulating substrate;
an immobilized enzyme layer formed over the ~~portion of~~ electrode;
an adhesion layer comprising a silane-containing compound formed over the immobilized enzyme layer; and
a permeation-limiting layer comprising a fluorine-containing polymer having a structure where a pendant group containing at least a fluoroalkylene block is attached to an unfluorinated vinyl-based polymer, which is formed on the adhesion layer,
wherein the fluorine-containing polymer contains no other fluorine than the fluorines contained in the fluoroalkylene block.
2. (original): An enzyme electrode as claimed in Claim 1 wherein said adhesion layer is a layer composed essentially of a silane coupling agent.
3. (currently amended): An enzyme electrode as claimed in Claim 1 or 2 wherein said fluorine-containing polymer is a fluoroalcohol ester of a polycarboxylic acid (A) in which

the fluoroalcohol portion is contained as the pendant group containing at least a fluoroalkylene block thereof, and the polycarboxylic acid (A) is contained as the unfluorinated vinyl-based polymer thereof.

4. (currently amended): An enzyme electrode as claimed in Claim 1 or 2 wherein said fluorine-containing polymer is a mixture that contains a fluoroalcohol ester of a polycarboxylic acid (A) in which the fluoroalcohol portion is contained as the pendant group containing at least a fluoroalkylene block thereof, and the polycarboxylic acid (A) is contained as the unfluorinated vinyl-based polymer and additionally an alkylalcohol ester of a polycarboxylic acid (B).

5. (currently amended): An enzyme electrode as claimed in Claim 1 wherein said fluorine-containing polymer is a copolymer composed of a fluoroalcohol ester of a polycarboxylic acid (A) and an alkylalcohol ester of a polycarboxylic acid (B),

in which the fluoroalcohol portion is contained as the pendant group containing at least a fluoroalkylene block thereof, and

the polycarboxylic acid (A) portion and the polycarboxylic acid (B) portion composes the unfluorinated vinyl-based polymer thereof.

6. (previously presented): An enzyme electrode as claimed in Claim 4 wherein said polycarboxylic acid (B) is selected from polymethacrylic acid, polyacrylic acid or a copolymer of acrylic acid and methacrylic acid.

7. (previously presented): An enzyme electrode as claimed in Claim 3 wherein said polycarboxylic acid (A) is selected from polymethacrylic acid, polyacrylic acid or a copolymer of acrylic acid and methacrylic acid.

8. (withdrawn – currently amended): An enzyme electrode comprising:
~~a portion of an~~ electrode formed on an insulating substrate;
an electrode protective layer covering the ~~portion of~~ electrode;
a binding layer comprising a silane-containing compound, which is formed on the electrode protective layer;
an ion-exchange resin film layer formed on the binding layer;
an immobilized enzyme layer formed on the ion-exchange resin film layer;
an adhesion layer comprising a silane-containing compound, which is formed on the immobilized enzyme layer; and
a permeation-limiting layer comprising a fluorine-containing polymer having a structure where a pendant group containing at least a fluoroalkylene block is attached to an unfluorinated vinyl-based polymer, which is formed on the adhesion layer,

wherein the fluorine-containing polymer contains no other fluorine than the fluorines contained in the fluoroalkylene block.

9. (withdrawn): An enzyme electrode as claimed in Claim 8 wherein said electrode protective layer is made essentially of a urea compound.

10. (withdrawn): An enzyme electrode as claimed in Claim 8 wherein said binding layer and said adhesion layer are layers composed essentially of a silane coupling agent.

11. (withdrawn – currently amended): An enzyme electrode as claimed in Claim 8 wherein said fluorine-containing polymer is a fluoroalcohol ester of a polycarboxylic acid (A) in which the fluoroalcohol portion is contained as the pendant group containing at least a fluoroalkylene block thereof, and the polycarboxylic acid (A) is contained as the unfluorinated vinyl-based polymer thereof.

12. (withdrawn – currently amended): An enzyme electrode as claimed in Claim 8 wherein said fluorine-containing polymer is a mixture that contains the fluoroalcohol ester of the polycarboxylic acid (A) in which the fluoroalcohol portion is contained as the pendant group containing at least a fluoroalkylene block thereof, and the polycarboxylic acid (A) is contained as the unfluorinated vinyl-based polymer and additionally an alkylalcohol ester of a polycarboxylic acid (B).

13. (withdrawn – currently amended): An enzyme electrode as claimed in ~~Claim 12~~ Claim 8, wherein said fluorine-containing polymer is a copolymer composed of a fluoroalcohol ester of a polycarboxylic acid (A) and an alkylalcohol ester of a polycarboxylic acid (B),
in which the fluoroalcohol portion is contained as the pendant group containing at least a fluoroalkylene block thereof, and
the polycarboxylic acid (A) portion and the polycarboxylic acid (B) portion composes the unfluorinated vinyl-based polymer thereof.

14. (withdrawn): An enzyme electrode as claimed in Claim 12 or 13 wherein said polycarboxylic acid (B) is selected from polymethacrylic acid, polyacrylic acid or a copolymer of acrylic acid and methacrylic acid.

15. (withdrawn): The enzyme electrode as claimed in any one of Claims 11 to 13 wherein said polycarboxylic acid (A) is selected from polymethacrylic acid, polyacrylic acid or a copolymer of acrylic acid and methacrylic acid.

16. (currently amended): An enzyme electrode comprising:
~~a portion of an~~ an electrode formed on an insulating substrate;
an immobilized enzyme layer formed on the ~~portion of~~ electrode;
an adhesion layer comprising a silane-containing compound, which is formed on the
immobilized enzyme layer; and

a permeation-limiting layer formed on the adhesion layer over the immobilized enzyme layer and placed on the uppermost surface;

wherein said permeation-limiting layer consists of a film essentially comprising a fluorine-containing polymer having a structure where a pendant group containing at least a fluoroalkylene block is attached to an unfluorinated vinyl-based polymer, and of which an average thickness of said permeation-limiting layer is selected within a range of 0.01 to 1 μ m; and many a plurality of grooves with depth selected in a range of 0.1 to 100 nm, which is due to variation of the thickness of said permeation-limiting layer, are built-present on the surface thereof of said permeation-limiting layer, and

the fluorine-containing polymer contains no other fluorine than the fluorines contained in the fluoroalkylene block.

17. (currently amended): An enzyme electrode as claimed in Claim 16 wherein an average thickness of said permeation-limiting layer is selected within a range of 0.02 to 0.5 μ m; and the surface of the permeation-limiting layer has an irregular shape having a surface roughness within a range of 0.0001 to 1, which surface roughness is defined as a ratio of mean value of the variation of the thickness to or more and 1 or less fold of said average thickness of the permeation-limiting layer.

18. (currently amended): An enzyme electrode as claimed in Claim 16 wherein said adhesion layer is a layer composed essentially of a silane coupling agent ~~said fluorine-containing~~

~~polymer is a polymer having a structure where a pendant group containing at least a fluoroalkylene block is attached to an unfluorinated vinyl-based polymer.~~

19. (currently amended): An enzyme electrode as claimed in Claim 16 or 18 wherein said fluorine-containing polymer is a fluoroalcohol ester of a polycarboxylic acid (A) in which the fluoroalcohol portion is contained as the pendant group containing at least a fluoroalkylene block thereof, and the polycarboxylic acid (A) is contained as the unfluorinated vinyl-based polymer thereof.

20. (currently amended): An enzyme electrode as claimed in Claim 16 or 18 wherein said fluorine-containing polymer is a mixture that contains the fluoroalcohol ester of the polycarboxylic acid (A) in which the fluoroalcohol portion is contained as the pendant group containing at least a fluoroalkylene block thereof, and the polycarboxylic acid (A) is contained as the unfluorinated vinyl-based polymer and additionally an alkylalcohol ester of a polycarboxylic acid (B).

21. (currently amended): An enzyme electrode as claimed in Claim 16 wherein said fluorine-containing polymer is a copolymer composed of a fluoroalcohol ester of a polycarboxylic acid (A) and an alkylalcohol ester of a polycarboxylic acid (B),

in which the fluoroalcohol portion is contained as the pendant group containing at least a fluoroalkylene block thereof, and

the polycarboxylic acid (A) portion and the polycarboxylic acid (B) portion composes the unfluorinated vinyl-based polymer thereof.

22. (previously presented): An enzyme electrode as claimed in Claim 20 wherein said polycarboxylic acid (B) is selected from polymethacrylic acid, polyacrylic acid or a copolymer of acrylic acid and methacrylic acid.

23. (previously presented): An enzyme electrode as claimed in Claim 19 wherein said polycarboxylic acid (A) is selected from polymethacrylic acid, polyacrylic acid or a copolymer of acrylic acid and methacrylic acid.

24. (withdrawn – currently amended): A process for manufacturing an enzyme electrode comprising the steps of:

forming an electrode film on the main surface of an insulating substrate and then patterning the electrode film to form a plurality of ~~portions of~~ electrodes;

forming an electrode protective layer covering the electrode surface;

forming a binding layer comprising a silane-containing compound on the main surface of the insulating substrate;

forming an ion-exchange resin film layer on the main surface of the insulating substrate;

applying a liquid containing an enzyme to the main surface of the insulating substrate and then drying the insulating substrate to form an immobilized enzyme layer;

applying a liquid comprising a silane-containing compound to the main surface of the insulating substrate and then drying the insulating substrate to form an adhesion layer, and sequentially applying a liquid containing a fluorine-containing polymer having a structure where a pendant group comprising at least a fluoroalkylene block is attached to an unfluorinated vinyl-based polymer to the upper surface of the adhesion layer coating the main surface of the insulating substrate by spin coating and then drying the insulating substrate to form the permeation-limiting layer; and

dicing the insulating substrate to give a plurality of enzyme electrodes;

wherein said permeation-limiting layer consists of a film comprising a fluorine-containing polymer having a structure where a pendant group containing at least a fluoroalkylene block is attached to an unfluorinated vinyl-based polymer.

an average thickness of said permeation-limiting layer is selected within a range of 0.01 to 1 μm ; and ~~many~~ a plurality of grooves with depth selected in a range of ~~0.1~~ 0.1 to 100 nm, which is due to variation of the thickness of said permeation-limiting layer, are built in present on the surface of the permeation-limiting layer, and

the fluorine-containing polymer contains no other fluorine than the fluorines contained in the fluoroalkylene block.

25. (withdrawn – currently amended): A process for manufacturing an enzyme electrode comprising the steps of:

forming an electrode film on the main surface of an insulating substrate and then patterning the electrode film to form a plurality of portions of electrodes;
forming an electrode protective layer covering the electrode surface;
forming a binding layer comprising a silane-containing compound on the main surface of the insulating substrate;
forming an ion-exchange resin film layer on the main surface of the insulating substrate;
applying an liquid containing an enzyme to the main surface of the insulating substrate and then drying the insulating substrate to form an immobilized enzyme layer;
applying a liquid comprising a silane-containing compound to the main surface of the insulating substrate and then drying the insulating substrate to form an adhesion layer, and sequentially applying a liquid containing a fluorine-containing polymer to the upper surface of the adhesion layer coating the main surface of the insulating substrate and then drying the insulating substrate to form a permeation-limiting layer; and
dicing the insulating substrate to give a plurality of enzyme electrodes,
wherein said permeation-limiting layer consists of a film comprising a fluorine-containing polymer having a structure where a pendant group containing at least a fluoroalkylene block is attached to an unfluorinated vinyl-based polymer, and
the fluorine-containing polymer contains no other fluorine than the fluorines contained in the fluoroalkylene block.

26. (withdrawn): A process for manufacturing an enzyme electrode as claimed in Claim 25 wherein said permeation-limiting layer is a layer being formed by spin coating.

27. (withdrawn): A process for manufacturing an enzyme electrode as claimed in Claim 25 wherein said silane-containing compound used for forming the adhesion layer is a silane coupling agent.

28. (currently amended): A biosensor comprising an enzyme electrode as claimed in ~~any one of Claims 1, 8 or 16~~ Claim 1 or 16.

29. (withdrawn – currently amended): A process for manufacturing an enzyme electrode as claimed in Claim 25 wherein said fluorine-containing polymer is a polymer having a structure where a pendant group containing at least a fluoroalkylene block is attached to an unfluorinated vinyl-based polymer, and

the fluorine-containing polymer contains no other fluorine than the fluorines contained in the fluoroalkylene block.

30. (withdrawn – currently amended): A process for manufacturing an enzyme electrode as claimed in Claim 25 or 29 wherein said fluorine-containing polymer is a fluoroalcohol ester of a polycarboxylic acid (A) in which the fluoroalcohol portion is contained

as the pendant group containing at least a fluoroalkylene block thereof, and the polycarboxylic acid (A) is contained as the unfluorinated vinyl-based polymer thereof.

31. (withdrawn – currently amended): A process for manufacturing an enzyme electrode as claimed in Claim 25 wherein said film comprising fluorine-containing polymer is a mixture that contains a fluoroalcohol ester of a polycarboxylic acid (A) in which the fluoroalcohol portion is contained as the pendant group containing at least a fluoroalkylene block thereof, and the polycarboxylic acid (A) is contained as the unfluorinated vinyl-based polymer and additionally an alkylalcohol ester of a polycarboxylic acid (B).

32. (withdrawn – currently amended): A process for manufacturing an enzyme electrode as claimed in Claim 25 wherein said fluorine-containing polymer is a copolymer of a fluoroalcohol ester of a polycarboxylic acid (A) and an alkylalcohol ester of a polycarboxylic acid (B),

in which the fluoroalcohol portion is contained as the pendant group containing at least a fluoroalkylene block thereof, and
the polycarboxylic acid (A) portion and the polycarboxylic acid (B) portion composes the unfluorinated vinyl-based polymer thereof.

33. (withdrawn – currently amended): A process for manufacturing an enzyme electrode as claimed in Claim 31 or 32 wherein said polycarboxylic acid (B) is selected from polymethacrylic acid, polyacrylic acid or a copolymer of acrylic acid and methacrylic acid.

34. (withdrawn): A process for manufacturing an enzyme electrode as claimed in Claim 30 wherein said polycarboxylic acid (A) is selected from polymethacrylic acid, polyacrylic acid or a copolymer of acrylic acid and methacrylic acid.

35. (new): An enzyme electrode as claimed in Claim 1,
wherein the adhesion layer comprising the silane-containing compound is formed by using a solution of the silane-containing compound in which a solvent used therein is chosen from the solvent group consisting of pure water and mixtures diluted with pure water of alcohols or esters in a final concentration of 5 % or less.

36. (new): An enzyme electrode as claimed in Claim 8,
wherein the adhesion layer comprising the silane-containing compound is formed by using a solution of the silane-containing compound in which a solvent used therein is chosen from the solvent group consisting of pure water and mixtures diluted with pure water of alcohols or esters in a final concentration of 5 % or less.

37. (new): An enzyme electrode as claimed in Claim 16,
wherein the adhesion layer comprising the silane-containing compound is formed by
using a solution of the silane-containing compound in which a solvent used therein is chosen
from the solvent group consisting of pure water and mixtures diluted with pure water of alcohols
or esters in a final concentration of 5 % or less.

38. (new): A process for manufacturing an enzyme electrode according to Claim 24,
wherein the liquid comprising the silane-containing compound is a solution of the silane-
containing compound in which a solvent used therein is chosen from the solvent group consisting
of pure water and mixtures diluted with pure water of alcohols or esters in a final concentration
of 5 % or less.

39. (new): A process for manufacturing an enzyme electrode according to Claim 25,
wherein the liquid comprising the silane-containing compound is a solution of the silane-
containing compound in which a solvent used therein is chosen from the solvent group consisting
of pure water and mixtures diluted with pure water of alcohols or esters in a final concentration
of 5 % or less.